

IRON IN THE AQUIFER SYSTEM OF SUFFOLK COUNTY, NEW YORK, 1990-98

ABSTRACT

igh concentrations of dissolved iron in ground water contribute to the biofouling of public-supply wells, and the treatment and remediation of biofouling are costly. Water companies on Long Island, N.Y., spend several million dollars annually to recondition,

redevelop, and replace supply wells and distribution lines; treat dissolved iron with sequestering agents or by filtration; and respond to iron-related complaints by customers. This report summarizes the results of studies done by the U.S. Geological Survey, in cooperation with the Suffolk County Water Authority, to characterize the geochemistry and microbiology of iron in the aquifer

system of Suffolk County. This information should be helpful for the siting and operation of supply wells.

Concentrations of dissolved iron in Long Island's ground water, and the frequency of iron biofouling of wells, are highest in ground-water-discharge zones, particularly near the south shore. Ground water along a deep north-south flowpath of the Magothy aquifer in southwestern

Figure 1. Encrusting material on a supply-well pump from Suffolk County, N.Y.

Suffolk County becomes anaerobic (oxygen deficient) and Fe(III) reducing at a distance of 8 to 10 kilometers south of the ground-water divide, and this change coincides with the downgradient increase in dissolved iron concentrations. The distribution of organic carbon, and the distribution and local variations in reactivity of Fe(III), in Magothy aquifer sediments have resulted in localized differences in redox microenvironments. For example, Fe(III)-reducing zones are associated with anaerobic conditions, where relatively large amounts of Fe(III) oxyhydroxide grain coatings are present, whereas sulfate-reducing zones are associated with lignite-rich lenses of silt and clay and appear to have developed in response to the depletion of available Fe(III) oxyhydroxides. The sulfatereducing zones are characterized by relatively low concentrations of dissolved iron (resulting from irondisulfide precipitation) and may be large enough to warrant watersupply development.

Specific-capacity and waterquality data from wells screened in the Magothy aquifer indicate that water from biofouled wells contains higher median concentrations of total and dissolved iron and manganese, total phosphate, and dissolved sulfate, and lower median concentrations of dissolved oxygen and alkalinity, and lower pH, than does water from unaffected wells. Corresponding data from wells screened in the upper glacial aquifer indicate that water from biofouled wells contains higher median concentrations of total and dissolved manganese and dissolved

sulfate, and lower pH, than does water from unaffected wells.

Filamentous bacteria were detected in 31 (or 72 percent) of the 43 biofilm samples obtained from biofouled wells during reconditioning. The predominant filamentous organism was Gallionella ferruginea, a major biofouling agent in the upper glacial and Magothy aquifers

throughout Suffolk County.
Mineral-saturation indices indicate that most of the well-encrusting material is deposited when the wells are shut down. Furthermore, the use of treated water (which has a high pH and sometimes high concentrations of dissolved iron) for pump prelubrication when wells are shut down could greatly increase the rate of iron oxidation.

INTRODUCTION

Dissolved iron in high concentrations is by far the most common water-quality problem associated with ground water (Chapelle, 1993) and can cause (1) staining and an unpleasant taste, (2) iron encrustation, which can decrease the specific capacity of wells and can clog pumps (fig. 1) and groundwater-treatment systems (Siegel and others, 1992), (3) anodic corrosion of steel pipes and water-distribution lines, and (4) acidification and Fe(III) hydroxide enrichment of soils (by oxidation of ferrous iron in irrigation water), which can severely diminish agricultural productivity (Chapelle, 1993). High concentrations of dissolved iron in ground water generally are caused by the microbial reduction of Fe(III), but also can be caused locally by the oxidation of iron disulfides.

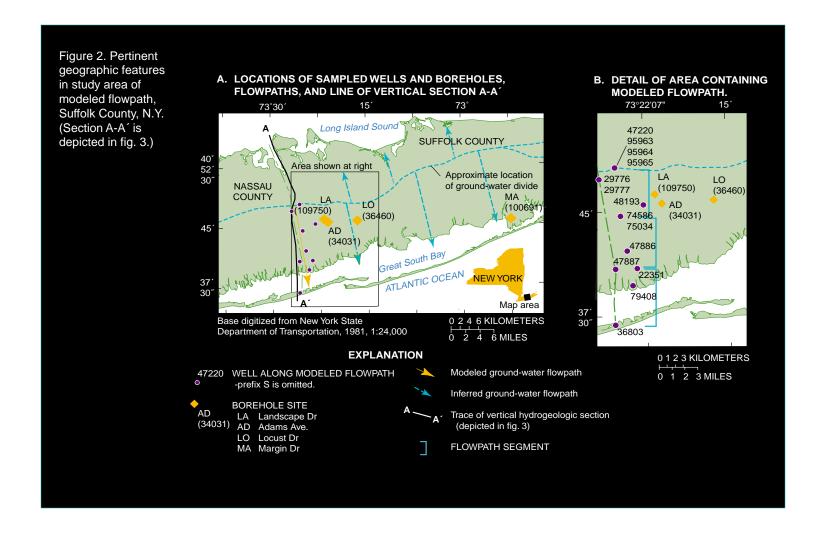
The term "aquifer biofouling" generally refers to the degradation of ground-water quality by bacteria and contributes to problems (2) and (3) listed above. Aquifer biofouling is probably the most costly iron-related problem in ground water (Smith, 1992) and is common in coastal and alluvial aquifers that contain high concentrations of dissolved iron (Applin and Zhao, 1989; Smith,

1992; Chapelle, 1993). Water companies on Long Island spend several million dollars annually in reconditioning, redeveloping, and replacing supply wells and distribution lines; treating dissolved iron with sequestering agents or by filtration; and responding to iron-related complaints by customers.

In 1990, the U.S. Geological Survey (USGS), in cooperation with Suffolk County Water Authority (SCWA), began a 3-year study to investigate the occurrence and process of well-screen encrustation through analyses of (1) chemical and hydrologic data from wells throughout the county, and (2) water samples and iron-biofilm samples from affected wells and sediment cores from selected sites (Walter, 1997a,b). Although much information on the biofouling process at the supply-well sites was obtained, uncertainty remained about the sources of iron, controls on iron solubility within the system, and geochemical changes along groundwater flowpaths.

In 1994, a 4-year followup study was begun to identify the sources and sinks of iron and other redox-active constituents along the southward flowpath from the ground-water divide to the southern shore at the Nassau-Suffolk County border, and to identify possible supply-well sites with minimal biofouling potential. The initial phase of the study entailed the delineation of (1) heavy-minerals distribution in the upper Cretaceous deposits (including the Magothy aquifer) and in the saturated part of the overlying Pleistocene deposits (upper glacial aquifer) through analysis of cores from boreholes, and (2) the distribution and morphology of iron-sulfide minerals at four new borehole sites in Suffolk County (Brown, 1998). The second phase of the study entailed geochemical and microbial characterization of iron in the Magothy aquifer (Brown, 1998; Brown and others, 1999). The uneven distribution of sediment texture, and of iron, sulfur, and carbon concentrations within the Magothy necessitated a detailed evaluation of core samples through analyses of pore water, aquifer mineralogy, and microbiology.

This report summarizes results of the two studies mentioned above; it also describes the hydrogeologic setting of the western Suffolk County study area, discusses cycling of iron within the aquifer system, and describes the biogeochemical aspects of biofouling.



HYDROGEOLOGIC SETTING

Long Island is underlain by a sequence of upper Cretaceous and Pleistocene sediments that were deposited on a southeastward dipping bedrock surface (figs. 2 and 3). The deposits thicken southeastward and reach a maximum thickness more than 600 m in southeastern Long Island. The principal aquifer on Long Island is the Magothy, which consists of the Matawan Group and Magothy Formation, undifferentiated, of Cretaceous age. Along the southern shore, the Magothy aquifer is unconformably overlain by the Monmouth Group (Monmouth greensand) of Cretaceous age;

elsewhere in the southern part of the island, it is unconformably overlain by the Gardiners Clay (an upper Pleistocene interglacial unit) and by glacial deposits of Pleistocene age (upper glacial aquifer). Cretaceous deposits north of the Gardiners Clay are unconformably overlain by the upper glacial aquifer (Smolensky and others, 1989).

Water in the upper glacial aquifer is generally oxic and contains low concentrations of dissolved iron and other cations, except in areas of discharge near the south shore, where the water has become anoxic through microbially mediated redox reactions along its flowpaths. The Magothy aquifer generally contains higher

concentrations of dissolved iron than does the upper glacial aquifer and, therefore, is subject to more pronounced iron biofouling.

FIELD AND LABORATORY METHODS

The initial iron study was conducted in Suffolk County during 1990-93 (Walter, 1997a,b); the followup study was done in Suffolk County and southwestern Nassau County in 1994-98 (Brown, 1998; Brown and others, 1999). The methods used for each study are described below.

Initial Iron Study (1990-93)

Water samples, biofilm samples, and sediment cores were collected from affected wells and test borings to identify the geochemical and microbiological mechanisms involved in the well-screenencrustation process. Locations of data-collection sites are given in Walter (1997b).

A total of 66 water samples were collected from 40 affected wells from January 1991 through August 1992. Additional water samples were collected from the lubrication systems of 16 affected wells. Treated water from the distribution system is trickled into the well during idle periods to keep pump turbines wet or

"lubricated." This process is referred to as "pump prelubrication." Water samples were collected before and after the reconditioning of 26 of the first 40 affected wells; an additional 14 samples were collected from wells that had histories of iron-related production declines but were not reconditioned during the study. A sufficient volume of water was evacuated after reconditioning to ensure that no residual effects of the reconditioning process remained. A detailed description of procedures used for water-sample collection and analyses are given in Walter (1997b).

Samples of encrusting material were collected from the pump

columns of 43 affected wells during reconditioning; the samples were collected with a sterilized metal scraper and distilled water as described in Walter (1997b). The material was analyzed with a light microscope to identify any filaments characteristic of iron-producing bacteria (Eleanor Robbins, U.S. Geological Survey, oral commun., 1990). Analyses of solid-phase material are described in Walter (1997b). In addition, a total of 118 sediment cores were collected from 7 replacement wells and 2 test borings to detect the presence of iron bacteria in the aguifer sediments, by methods described in Walter (1997b).

Followup Iron Study (1994-98)

Water samples were collected from monitoring wells and publicsupply wells along a Magothy aquifer flowpath near the Nassau-Suffolk County border. Procedures for the collection and analyses of waterchemistry data are described in Brown and others (1999).

Split-spoon cores of Magothy sediment were obtained from four boreholes (fig. 2) by a reverse-rotary drill rig, and several cores from each borehole were selected for analysis. The cores were designated as follows:

- AD1 through AD5 (Adams Avenue).
- LA1 through LA3 (Landscape Drive),
- LO1 through LO6 (Locust Drive), and
- MA1 through MA4 (Margin Drive) (locations are shown in fig. 2).
 Subsamples of sediment cores from several depths were examined for

the predominant anaerobic microbial populations at several depths through terminal-electronaccepting-process (TEAP)identification assays as described in Brown and others (1999). Ferric iron coatings on sediments were measured by 0.5 M HCl and 6 M hydroxylamine extraction as described in Lovley and Phillips (1987). Core samples generally yielded insufficient pore water for analysis; therefore, a subsampled part of the core was diluted with deionized water, followed by filter pressing with N2 (Lusczynski, 1961). A gamma log, lithologic description, and depth profile of organic carbon, total sulfur, anaerobic bacteria, and iron coatings in sediments, and concentrations of dissolved iron, sulfate, and dissolved organic carbon (DOC) in pore water, are

given in Brown and others (1999). A detailed description of methods for the collection and analyses of core samples are given in previous reports (Brown and others, 1999; Brown, 1998).

Laboratory procedures for study of core-sample mineralogy entailed grain-size analysis, heavy-mineral separation, element analysis with energy-dispersive spectroscopy on an electron microprobe analyzer, and powder x-ray diffraction on a Scintag diffractometer. Scanning electron microscopy (SEM) was used to identify crystal morphology.

Ion-exchange experiments were conducted under anoxic conditions in a glove box with nitrogen gas as described in Brown (1998). Ion exchange in the Magothy aquifer was modeled with the computer code PHREEQM (Appelo and Postma, 1993), as described in Brown (1998).

IRON IN THE AQUIFER SYSTEM OF SUFFOLK COUNTY

Dissolved iron represents only a small part of the total iron within an aquifer (Heron and Christensen, 1995); the rest is bound in ironbearing minerals. Previous studies on Long Island have shown that the spatial distribution, and the reactivity, of iron-bearing minerals in the Pleistocene and upper Cretaceous deposits vary with the predominant terminal-electronaccepting process (TEAP) zones in the aquifer, which in turn are a function of the depth and distance from the ground-water divide, the organic-matter content of the sediment, and the type of depositional environment (Brown, 1996; Brown, 1998). The most abundant and potentially reactive of the iron-bearing mineral grains were found to be iron (hydr)oxides, leucoxene, glauconite, chlorite, pyrite, and marcasite. Fe(III) oxyhydroxide grain coatings, which are probably the most microbially reducible iron phase in the aquifer system, are discussed in Brown and others (1999).

Iron Reactions

Redox reactions in groundwater systems commonly occur as a sequence of TEAPs along a flowpath (Champ and others, 1979; Jackson and Patterson, 1982). Reduction of electron acceptors along ground-water flowpaths typically proceeds from highest to lowest energy yield, beginning with the reduction of oxygen, followed by reduction of nitrate, Mn(IV), Fe(III), sulfate, and carbon dioxide. The oxidation of

organic matter by fermentative bacteria produces simple organic acids and H₂, which are required by anaerobic bacteria for reduction of electron acceptors (Lovley and Goodwin, 1988; Lovley and others, 1990). Ground water along the Magothy flowpath (fig. 2) becomes anaerobic and Fe(III) reducing at a distance of 8 to 10 km south of the ground-water divide, as indicated by the downgradient decrease in dissolved oxygen (fig. 4A) and increase in dissolved iron (fig. 4B) concentrations. Reduction of nitrate (denitrification) probably occurs before the reduction of Fe(III), but the initial nitrate concentrations generally are low (< 0.004 to 0.16 mmol/L). Mn(IV) is reduced after NO3- and before Fe(III). Although the spatial distribution of dissolved manganese concentrations in wells of Suffolk County is similar to that of iron concentrations (Walter, 1997a), the manganese concentrations are lower and reflect the lower concentrations in sediment-grain coatings (Brown, 1998). Dissolved inorganic carbon (DIC) concentrations (fig. 4D) and pH also increase southward along the flowpath. Mass-balance modeling along flowpath segments between representative wells along the flowpath (fig. 4) was done to estimate the mass transfer of redoxactive constituents, as described in Brown (1998).

The chemical trends shown in figure 4 represent ground water from Magothy wells and generally do not reflect conditions observed in poorly permeable sediments from core samples; the dashed red lines connect representative ground-water samples from wells that were used in the geochemical-modeling study (Brown, 1998). The availability and distribution of organic carbon in

Magothy sediments, and the locally varying distribution and reactivity of Fe(III), have resulted in localized redox microenvironments (Brown and others, 1999). The variation in chemical concentrations around the trend lines in figures 4C and 4D reflect the uneven distribution of sulfate and organic carbon in aquifer sediments. High DIC concentrations near the ground-water divide at wells 95963 and 34031 (fig. 4D) probably reflect the (1) liming of home lawns, cemeteries, and parks, and (or) (2) contribution from septic systems. The microbial and geochemical analyses of core samples from the four SCWA borehole sites (fig. 2A) indicate that the predominant anaerobic redox process in the aquifer is sulfate reduction, which occurs in localized zones associated with poorly permeable lignitic sediments, high concentrations of dissolved organic carbon and sulfate, and low concentrations of ferric iron grain coatings (fig. 5). The redox processes were identified primarily through microbial analysis of core samples.

Sediment cores from along the anaerobic part of the modeled flowpath (fig. 2) were unavailable for TEAP identification, but H₂ concentrations in ground water from wells in southwestern Suffolk County, and results of geochemical flowpath modeling, confirm the predominance of sulfate reduction (Brown and others, 1999). Low aqueous-sulfide concentrations (<0.25 to 0.7 μ mol/L as H₂S) in water from the anaerobic part of the flowpath indicate that Fe(II) in the water reacts with sulfide to form pyrite or marcasite (FeS₂). The predominant redox process in sandy sediments with less lignite was Fe(III) reduction (fig. 5); a unique iron-reducing microorganism (MD-612) was found and isolated from

sediments at the Margin Dr. site in south-central Suffolk County (Brown and others, 1999). The localized sulfate-reducing zones, which appear to have developed in response to the depletion of available Fe(III) oxyhydroxides, contain relatively low concentrations of dissolved iron as a result of the precipitation of iron-sulfide minerals. Furthermore, these sulfate-reducing zones may be large enough to be suitable for water-supply development (Brown and others, 1999).

Iron-sulfide minerals represent a large source and sink of iron in the Magothy aquifer. The differing lithologic occurrences of pyrite and marcasite (FeS₂), and their wide ranges of morphology (fig. 6), indicate that chemical conditions (for example, redox, pH, and concentrations of sulfate and ferric oxyhydroxides) vary spatially and

temporally (Brown, 1998). Pyrite was generally found in association with lignite (figs. 6A, 6B) or as interstitial cement. Marcasite was found as interstitial cement (fig. 6C) and is associated with the oxidation of upgradient FeS₂ (fig. 5D).

Sulfate concentrations also increase downgradient (fig. 4C), despite the predominance of sulfatereducing TEAPs along most of the flowpath. The increase in sulfate concentrations southward along the flowpath in western Suffolk County, and the high concentrations observed in pore waters extracted from core samples, are consistent with observations made in other studies in Atlantic Coastal Plain aquifers (summarized by Chapelle, 1993). Although several sources and mechanisms of high sulfate concentrations in ground water have been proposed, their origin is poorly

understood. Some of the sulfate is reduced by microorganisms in the Magothy aquifer, where it reacts with iron to form iron-sulfide minerals (Brown and others, 1999). Identifying the source of sulfur, therefore, is important because it indirectly affects the mobility of iron. Ion-exchange experiments indicate that sorbed sulfate, which could be of marine origin and related to a sea-water transgression, is small but significant in the acidic pore water of poorly permeable clays (Brown, 1998) and can contribute to the downgradient increase of sulfate concentration in ground water. Some of the sulfate released can result from oxidation of FeS₂ by aqueous Fe(III) under anoxic conditions, as evidenced by the ionexchange experiments, as well as by etched FeS₂ samples and in d34S trends along the flowpath (Brown, 1998). This oxidation of FeS₂ can

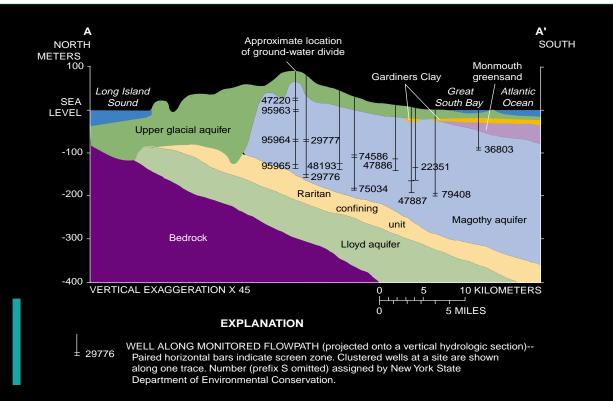


Figure 3. Hydrogeologic section A-A´ and sampled wells along Nassau-Suffolk County border, N.Y. (Modified from Smolensky and others, 1989, sheet 1.)

Figure 4.
Concentrations of dissolved (A) oxygen, (B) iron, (C) sulfate, and (D) inorganic carbon, in ground water along a flowpath in the Magothy aquifer at the Nassau-Suffolk County border, Long Island, N.Y.

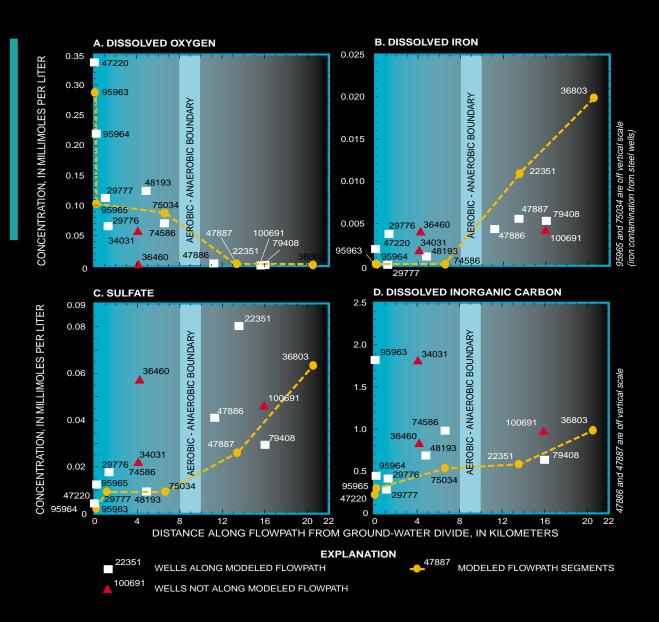
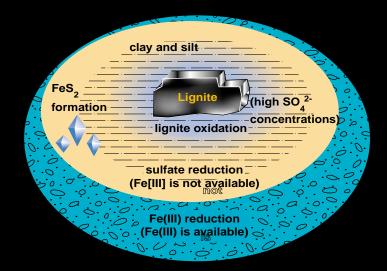
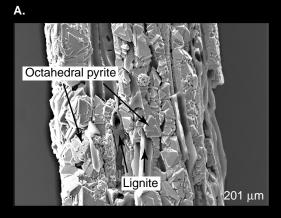
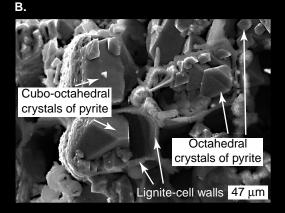
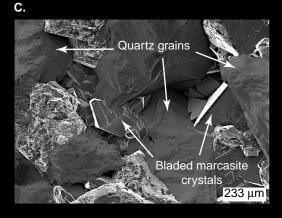


Figure 5. Geochemical and microbially mediated reactions that are potentially associated with lignitic microenvironments in the Magothy aquifer, Suffolk County, N.Y. These microenvironments were also found in aerobic parts of the aquifer; pyrite is oxidized at the perimeter of these microenvironments.









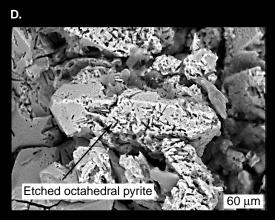


Figure 6. Scanningelectron microscope images of iron-sulfide minerals showing (A) octahedral pyrite crystals on lignite at site MA, (B) pyrite crystals and lignite wood-cell structures at site LO, (C) platey crystals of marcasite cement at site AD, and (D) etched pyrite at site AD. (Site locations are shown in fig. 2.)

also contribute to the downgradient sulfate increase in ground water.

The small-scale microbial and geochemical differences observed among the core samples are not typically evident at the regional scale represented by ground-water samples from wells. The generalized, regional approach identifies the major TEAPs but not the localized zones of microbial and geochemical reactions, which can be important for assessing zones of low dissolved iron concentration for water-supply development.

Iron-related biofouling

Aquifer biofouling is first stimulated by the redox gradient created near the well screen by the introduction of oxygen when the well is drilled and operated in an anaerobic aquifer. Aerobic organisms grow at this redox gradient as a layer, generally called a "biofilm," on the pump, well screen, and surrounding sediments. The biofilm then becomes stratified into a relatively thin layer of aerobic bacteria overlying a thicker anaerobic zone (Cullimore, 1986) and provides a medium for the growth of filamentous iron bacteria such as Gallionella. The biofilm also grows in volume and obstructs aguifer pore spaces and the well screen and, consequently, lowers the well's yield and specific capacity. (Yield is defined as the discharge, in units of volume per time unit; specific capacity is defined as the yield divided by the change in head.) The biomass around the supply-well screen acts as a protective coating for the iron-related bacteria within, and together these cause changes in water quality, including the removal of iron, manganese, and nutrients. Biofilms also protect bacteria cells within them from external stressors such as disinfectants (Smith, 1992).

Consequently, water from the supply well can differ chemically from ambient ground water (Cullimore, 1986). Samples from "outpost" monitoring wells, which do not have the high yields that introduce oxygen and promote growth of the biomass, can be more representative of aquifer chemistry than those from the supply well.

A correlation between declining specific capacity and high iron concentrations was noted by Walter (1997a). Water from Magothy wells with annual specific-capacity decreases of 5 percent or more had higher concentrations of iron, manganese, total phosphate, and dissolved sulfate, and lower concentrations of dissolved oxygen, nitrate, alkalinity, and lower pH, than unaffected Magothy wells. Data from the upper glacial aquifer indicate higher median concentrations of manganese and sulfate, and lower pH, in biofouled wells than in unaffected wells. The high percentage of biofouled wells in ground-waterdischarge zones, where reducing conditions prevail, indicates a correlation between specific-capacity decreases and the presence of chemical constituents associated with reducing environments.

Some wells that are screened in similar environments can differ in their symptoms of biofouling as a result of variations in well operation or design. Cyclic pumping or long periods of idleness, for example, can promote biofilm growth and associated problems (Smith, 1992). Mineral-saturation indices and ironoxidation kinetics in water samples from wells in Suffolk County indicate that most of the well-encrusting material was deposited when the wells were shut down (Walter, 1997b). Furthermore, lubricating the wells with treated water, which has a

high pH and sometimes high concentrations of dissolved iron, could greatly increase the rate of iron oxidation (Walter, 1997b).

Gallionella ferruginea is the predominant filamentous organism and is a major biofouling agent in Suffolk County (Walter, 1997b). This species requires oxygen but prefers water with low concentrations of dissolved oxygen and high concentrations of dissolved iron. Biofilm samples were obtained from the encrusting material of 43 affected wells during reconditioning. Much of the encrusting material (fig. 1) consisted of extracellular bacteria filaments, and the remainder was primarily amorphous ferric hydroxide with lesser amounts of goethite, hematite, other iron oxides, and silica. Filamentous bacteria were observed in samples from 31 of the 43 affected wells. The detection of Gallionella ferruginea in all biofilm samples collected from 12 locations within the SCWA distribution system indicates that Gallionella is likely to be present in all distribution and circulation water. The detection of ironbiofouling bacteria in core samples from several locations within the aquifer system in Suffolk County indicates their natural presence in the aquifer. Iron bacteria are transported through the ground-water system as small (0.1 mm to 0.3 mm diameter), inactive cells that attach themselves to surfaces and begin to grow when conditions become favorable, such as near a well screen (Cullimore, 1986). Iron-related bacteria have been found to be highly resilient and pervasive throughout aquifer systems (Cullimore, 1986). As a result, approaches to eliminate the bacteria problem are limited, and emphasis is placed instead on preventive treatment with disinfectants, acids, and surfactants (Hackett, 1987).

CONCLUSIONS

High concentrations of dissolved iron in ground water of Suffolk County cause staining and bad taste and promote the growth of ironbiofouling bacteria and iron encrustation around the screens of production wells, decreasing their specific capacity. Treatment of water and maintenance of wells and distribution lines is costly to water suppliers. The extent of iron encrustation and aquifer biofouling of supply wells is affected by (1) iron and redox chemistry, which differs between aquifers and varies locally throughout Long Island, (2) placement of wells, and (3) pumping practices.

Ground water along a deep flowpath in the Magothy aquifer in southwestern Suffolk County becomes anaerobic and Fe(III) reducing at a distance of 8 to 10 km south of the ground-water divide. This change corresponds to the downgradient increase in dissolved iron concentrations. The availability and distribution of organic carbon, together with the varying distribution and reactivity of Fe(III) in Magothy sediments, has resulted in localized differences in redox microenvironments. Fe(III)-reducing zones are associated with anaerobic conditions where Fe(III) oxyhydroxide grain coatings are prevalent. In contrast, localized sulfate-reducing zones are associated with lignite-rich, silt-and-clay lenses and appear to have developed in response to the depletion of available Fe(III) oxyhydroxides. The sulfatereducing zones are characterized by somewhat lower concentrations of dissolved iron because of iron-sulfide mineral precipitation, and these zones may be large enough for watersupply development.

The high percentage of biofouled wells in ground-water-discharge zones near the south shore of Long Island, where reducing conditions prevail, indicates a correlation between specific-capacity decreases and the concentrations of chemical constituents associated with reducing environments. Ground water from biofouled wells screened in the Magothy aquifer contains higher median concentrations of total and dissolved iron and manganese, total phosphate, and dissolved sulfate, and lower median concentrations of dissolved oxygen and alkalinity, and lower pH, than ground water from unaffected Magothy wells. Ground water from biofouled wells in the upper glacial aquifer contains higher median concentrations of total and dissolved manganese and dissolved sulfate, and lower pH, than does ground water from unaffected wells.

Of 43 biofilm samples obtained from the encrusting material of affected wells during reconditioning, 31 (72 percent) contained filamentous bacteria. Gallionella ferruginea was the predominant filamentous organism and is a major biofouling agent in the upper glacial and Magothy aquifers of Suffolk County. Mineral-saturation indices indicate that most of the well-encrusting material is deposited when the wells are shut down. Furthermore, the lubrication of pumps with treated water, which has a high pH and sometimes high concentrations of dissolved iron, could greatly increase the rate of iron oxidation.

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